Global aerosol modeling with the online multiscale NMMB/BSC Chemical Transport Model: sensitivity to secondary organic aerosol schemes (preliminary results)

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INTRODUCTION

The state-of-the-art of global modeling of organic aerosols is characterized by uncertainties of one order of magnitude (or more) in terms of primary production, secondary formation, surface concentrations, and vertical distribution (Tsigaridis et al., 2014). One of the main source of models' diversity is represented by the complexity of secondary organic aerosols (SOA) parameterization. However, the AEROCOM intercomparison study of Tsigaridis et al. (2014) (involving 31 CTMs and GCMs) showed that there is not a clear improvement when the model complexity is increased.
In this work, we focus on the complexity of the biogenic SOA formation parameterization by investigating the influence of using a 2-product scheme instead of a prescribed conversion of gaseous emissions.

METHODS

We implement two different SOA parameterizations in the BSC Chemical Transport Model aerosol module (Spada et al., in prep.), which is online coupled with the Non-hydrostatic Multiscale Model (Janjic et al., 2011; Janjic and Gall, 2012) (NMMB/BSC-CTM).

In the first experiment (EXP1), we consider only 1 hydrophilic SOA tracer and we assume a very simplified production of SOA by a constant-yield conversion of monoterpenes' emissions flux (Dentener et al., 2006):

$$F^{(SOA)} = 0.15 \cdot F^{(TERP)}$$

In the second experiment (EXP2), we transport 4 hydrophilic SOA tracers and we use a 2-product SOA scheme based on terpenes (TERP) and isoprene (ISOP) gas-phase oxidations (Tsigaridis and Kanakidou, 2007):

ISOP(g) + OH(g) → ISOP-P1(g) + …
ISOP(g) + O3(g) → ISOP-P2(g) + …
TERP(g) + OH(g) → TERP-P1(g) + …
TERPg) + O3(g) → TERP-P2(g) + …

Then gas/particle partitioning is applied.
Terpenes and isoprene emissions are provided by the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2006), To obtain organic aerosol concentrations, the 2 primary organic aerosol tracers transported by the model are added to SOA tracers. Anthropogenic emissions are from the AEROCOM-ACCMIP dataset and the fires' injection height is according to the IS4FIRES satellite-derived climatologies (Sofiev et al., 2012). We simulated a 5-year period (2002 – 2006) at 1 degree of horizontal resolution. An OA/OC ratio of 1.8 is applied.

PRELIMINARY RESULTS

We investigate the differences introduced by the SOA parameterizations in EXP1 and EXP2 on the monthly mean surface concentrations of organic carbon at global scale (annual mean values shown in Fig. 1). Our preliminar results indicate that there is a significant bias (up to a factor of 2) between the two experiments; in particular, the SOA concentrations simulated by EXP1 are strongly reduced in Amazonia, Australia, New Zealand, and high-latitude North America (Fig. 2) when the EXP2 scheme is applied.
However, despite the relevance of bias, the correlation between the seasonality simulated by EXP1 and EXP2 does not significantly change globally (Fig. 3); the only large area affected by significant changes is the Northern Russia ($r<0.5$). These results are mainly due to the combined action of terpenes and isoprene of EXP2. With the exception of high-latitudes, the isoprene seasonality is highly correlated with the terpenes' one. Then, the sum of terpenes and isoprene contributions can be reproduced by a terpene-like behavior, which is the case of EXP1 (with a tuning factor of 0.15). A significant exception to this behavior can be found in the Northern Russia, where the reduced isoprene contribution to SOA in the boreal summer is compensating the monoterpenes' maximum.
A global comparison with observations is very uncertain due to the lack of globally distributed measurements. The most comprehensive mass concentration measurement network is provided by IMPROVE over USA, where EXP1 and EXP2 show minimal differences between them (see Colorado station shown in Fig. 2). Few other worldwide measurement sites are availables, such as Manaus station (shown in Fig. 2), but they are not sufficient to give a clear evaluation of the EXP2-EXP1 bias and seasonality. No measurements are available in the Northern Russian region to compare with the marked changes in the simulated seasonality (see the plot in Fig. 3). Overall, our preliminar results point out that the upgrade from a prescribed constant yield scheme to a 2-product scheme of biogenic SOA provides significant changes in the global simulation of absolute values of organic carbon concentrations, but not in their seasonality.

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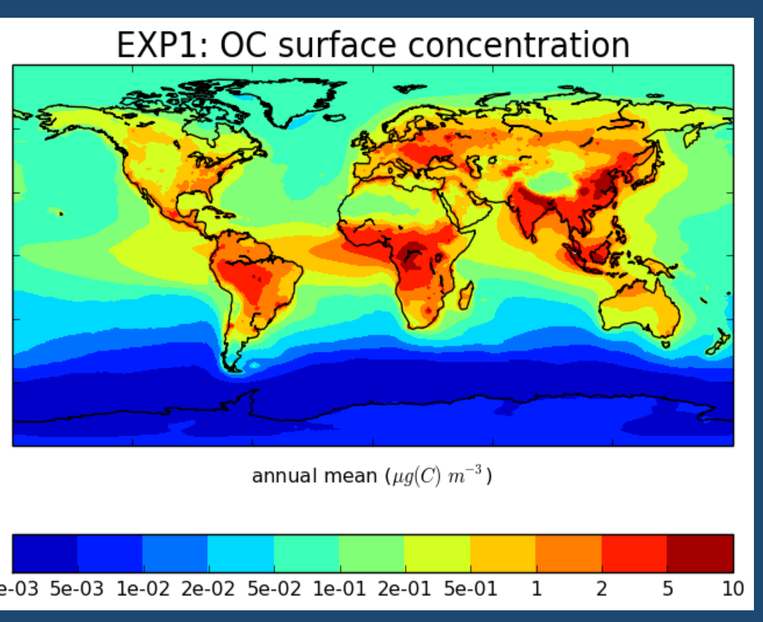
Figure 1



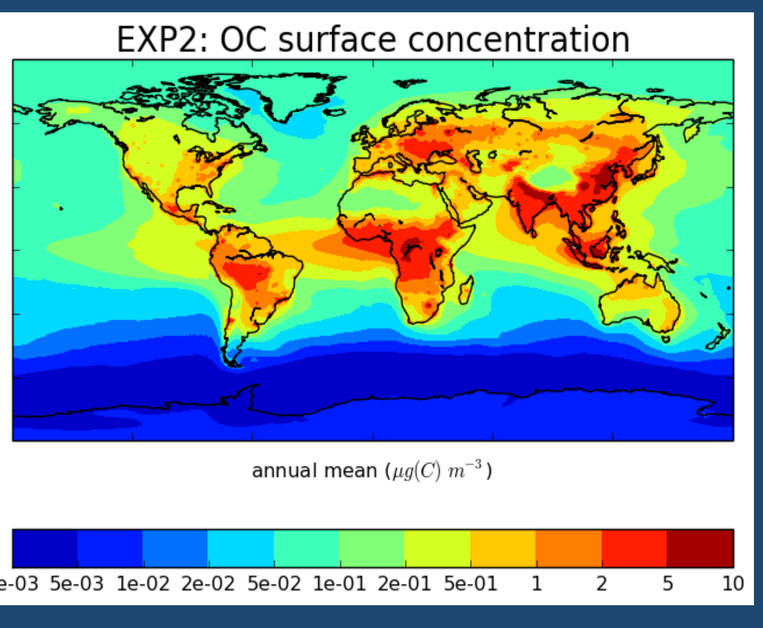
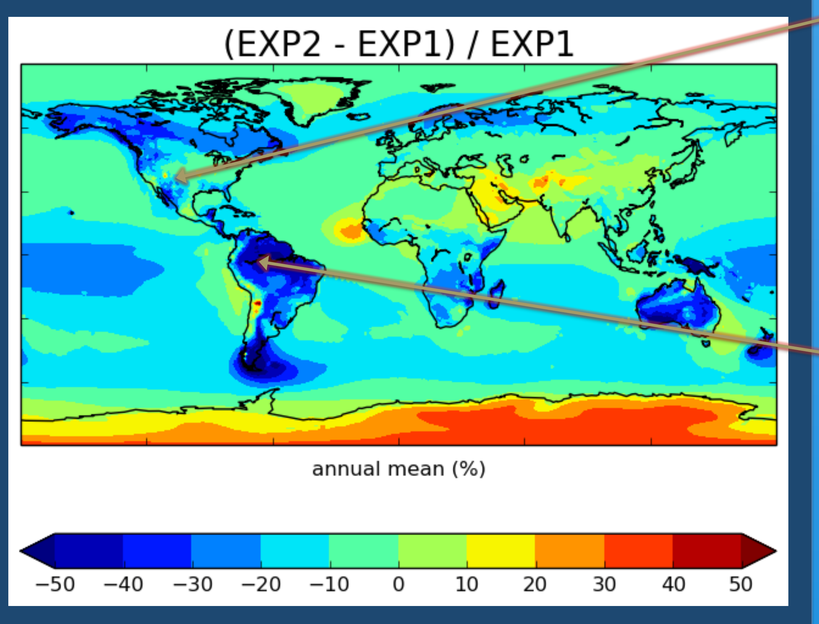
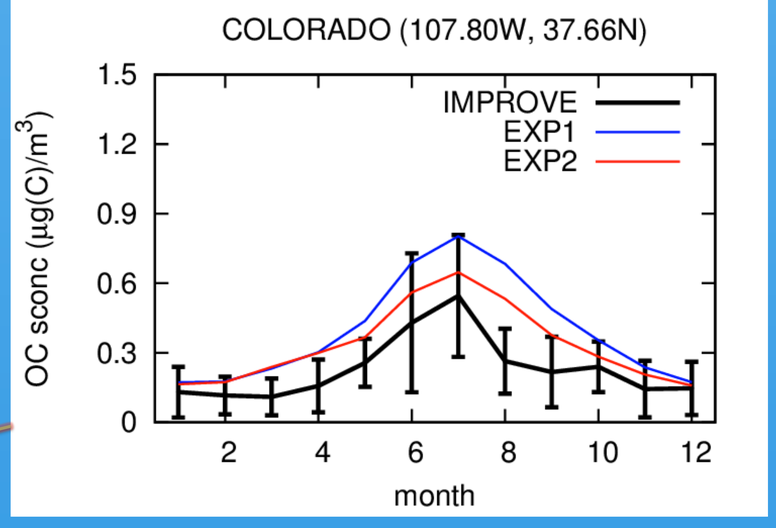


Figure 2





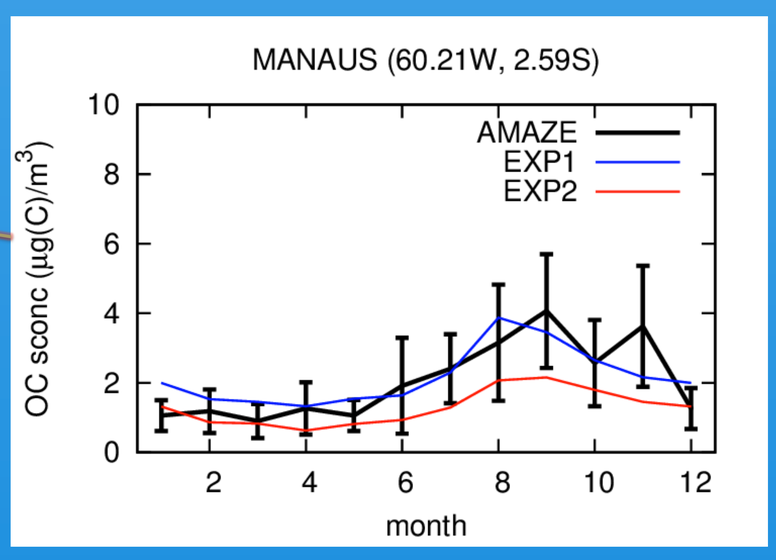
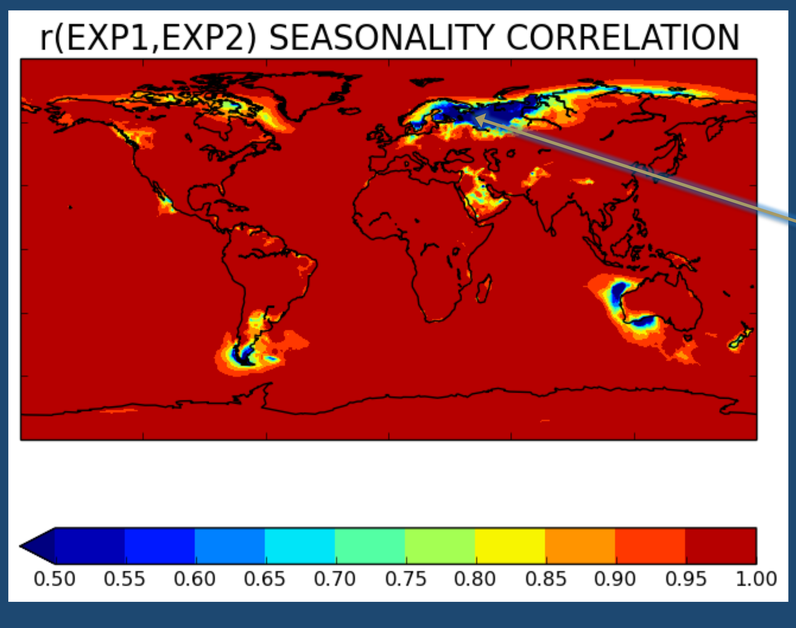
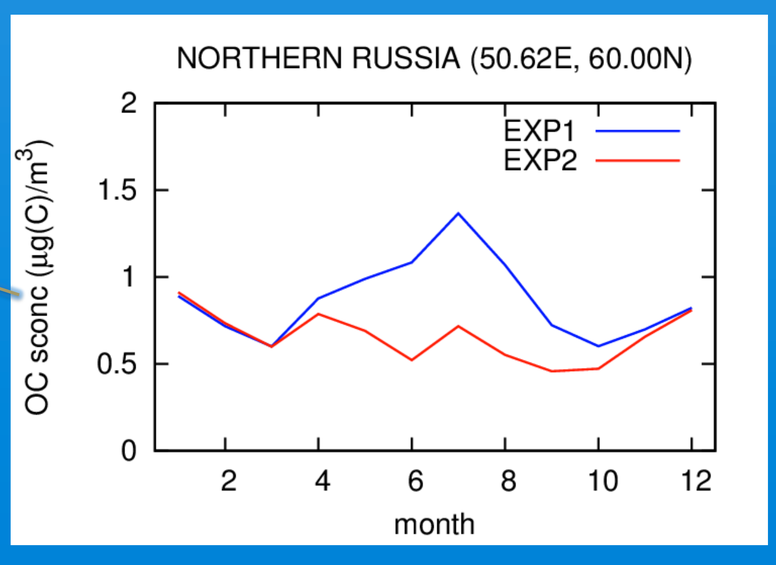


Figure 3





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